Risk assessment of GenX and PFOA in vegetable crops in Dordrecht, Papendrecht and Sliedrecht

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M.J.B. Mengelers (author), RIVM
J.D. in Biesebeek (author), RIVM
M. Schipper (author), RIVM
W. Slob (author), RIVM
P.E. Boon (author), RIVM

Contact:
Marcel Mengelers VPZ / VVH
marcel.mengelers@rivm.nl

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PO Box 1 | 3720 BA Bilthoven The Netherlands
www.rivm.nl

Citizens' summary

Risk assessment of GenX and PFOA in vegetable crops in Dordrecht, Papendrecht and Sliedrecht

Due to the emissions of the chemistry company DuPont / Chemours in Dordrecht, the substances GenX and PFOA have been released into the environment via the air. As a result, people with a vegetable garden near the company wonder whether it is safe to eat home-grown vegetables. The limit values that apply to the exposure of GenX and PFOA are not exceeded via food, according to research by the RIVM. Local residents also come into contact with the substances through air and drinking water. Therefore, the RIVM advises to consume vegetable crops that have been grown within a radius of 1 kilometer from the farm in moderation (not too often or too much). There, somewhat higher concentrations were found. Outside this area, concentrations are so low that the crops can be safely eaten, also in combination with other exposure sources.

The basis of this research is the calculation of the exposure to GenX and PFOA via home-grown vegetables in people with a vegetable garden within a radius of 4 kilometers around the factory. At the end of August 2017 samples of vegetables were taken at 10 locations: in Dordrecht and Papendrecht at three locations and at four in Sliedrecht. As a comparison, a location in Bilthoven was investigated. At all locations, samples were taken from three categories of vegetables (leaf, tuber and fruit vegetables). Two fruit plants were also examined at one of the locations. A total of 81 samples were analyzed.

GenX and / or PFOA have been demonstrated in about 40 percent of the samples around the plant. GenX was found in 14 percent of the samples around the plant in measurable quantities and in 4 percent PFOA. At concentrations lower than 1 nanogram per gram, the precise quantity can not be indicated; only the observation that it is in it. At one location, less than 1 kilometer northeast of the plant, higher concentrations of GenX were found in vegetables (in endive, beet, celery, lettuce and tomatoes) and PFOA (in beet) than in the other 9 locations around the plant.

The highest concentrations were then used to calculate the exposure. It is assumed that people will only eat vegetables from their own garden every day. The results are therefore probably higher than the actual exposure of GenX and PFOA at vegetable gardeners around the factory. Under these worst-case conditions, the exposure of both substances via food did not exceed the limit values that are considered safe (health-based limit values).

Key words: GenX, PFOA, risk assessment, vegetable garden, fruit and vegetables

Synopsis

Risk assessment of GenX and PFOA in vegetable garden crops in Dordrecht, Papendrecht and Sliedrecht

As a result of DuPont / chemicals chemicals company in Dordrecht, the substances GenX and PFOA have emitted into the environment via the air. As a consequence, people with a vegetable garden in the vicinity of the company are not sure whether it is safe to eat their home-grown vegetables. A study carried out by RIVM concludes that the threshold values of GenX and PFOA are not exceeded. However, residents are also exposed to these substances via air and drinking water. RIVM therefore advises that vegetables grown grown within a radius of 1 kilometer from the company should be consumed in moderation (not too often or too much). The concentrations found within this area were somewhat higher. Outside this area, the concentrations were so low that the crops can be safely consumed if one takes in account the other sources of exposure.

The study is based on GenX and PFOA through home-grown vegetables with a vegetable garden located within radius or 4 kilometers from the factory. At the end of August 2017, samples were tasks of vegetables at different locations, at three locations in Dordrecht and Papendrecht and at four locations in Sliedrecht. A location in Bilthoven was sampled for purposes of comparison. Samples were tasks or three categories of vegetables at all locations, namely leafy vegetables, root vegetables, and fruiting vegetables. At one of the locations, two fruit crops were also sampled. A total of 81 samples were analyzed.

GenX and / or PFOA were found in about 40% of the samples at the vicinity of the factory. GenX was found to be present in measurable quantities in 14% of the samples in the vicinity of the factory, and the same was true or PFOA in 4% of the samples. At concentrations less than 1 nanogram per gram, it was not possible to determine the exact quantity present, but only that the substance was present. At one location, less than 1 kilometre north-east of the factory, higher concentrations of GenX (in endive, beets, celery, lettuce, and tomatoes) and of PFOA (in beets) were found in vegetables than at the other nine locations close to the factory.

The highest concentrations were used to calculate the exposure. The calculation was based on the assumption that the persons would live in their lives. The calculated results are therefore probably higher than the actual exposure to GenX and PFOA or vegetable garden owners in the vicinity of the factory. Under these worst-case circumstances, the exposure to both substances via food did not exceed the threshold values that are considered safe (the so-called health-based guidance values).

Keywords: GenX, PFOA, risk assessment, home-grown, vegetables and fruit

Index

Resume

The Vrije Universiteit Amsterdam (VU) conducted an exploratory study in 2017 into the GenX and PFOA levels in grass and leaves in the vicinity of the DuPont / Chemours company. As a result, people with a vegetable garden near the company wonder whether it is safe to eat home-grown vegetables. However, the RIVM was unable to make any pronouncement on the basis of the results of the VU. That is why the Municipality of Dordrecht, for the cooperating authorities (Sliedrecht, Papendrecht, Province of South Holland and the Government) asked the RIVM to conduct research in vegetable gardens in the vicinity of DuPont / Chemours.

The aim of this research was to find out whether people can safely eat the vegetables from a vegetable garden in the vicinity of DuPont / Chemours with regard to the presence of GenX and PFOA in the vegetables. To this end, two specific research questions have been asked:
1. What are the concentrations of GenX and PFOA in selected crops from vegetable gardens in the vicinity of DuPont / Chemours?
2. Is the allowable daily intake (TDI) via food from GenX and PFOA exceeded by consumption of vegetable crops in a typical consumption pattern?

At the end of August 2017, samples of vegetables were taken at 10 locations in the vicinity of the Dupont / Chemours plant: three locations in Dordrecht, three in Papendrecht and four in Sliedrecht.
In addition, a reference site in Bilthoven has been sampled. At all locations, samples were taken from three categories of vegetables (leaf, tuber and fruit vegetables). Two fruit plants were also examined at one of the locations. A total of 81 samples were analyzed by the RIKILT research institute of the University of Wageningen for the presence of GenX and PFOA.

To carry out a risk assessment of GenX and PFOA present in vegetables and fruit in vegetable gardens, the intake of these substances was compared with the health-based limit values (allowable daily intake, TDI) of GenX and PFOA. For the calculation of intake, the measured concentrations of GenX and PFOA in the samples were combined with consumption quantities of vegetables and fruit from two Dutch food consumption surveys. The TDIs for GenX and PFOA have been derived by the RIVM in the recent past. When comparing the intake and the TDI of both substances, the uncertainties in the available data were taken into account in the interpretation where possible.

The study of the concentrations of GenX and PFOA in the vegetable crops yielded, in summary, the following results. At the reference site, GenX or PFOA were not detected in any of the analyzed crops (seven samples of vegetables). GenX and / or PFOA were detected in approximately 40% of the remaining 74 samples taken in the vicinity of the DuPont / Chemours plant. One location, less than 1 km northeast of DuPont / Chemours, showed higher concentrations of GenX and PFOA than the other nine locations. GenX was quantified in five crops (endive, beet, celery, lettuce and tomatoes) and the highest average concentration was 5.4 nanograms of GenX / gram of lettuce. PFOA was quantified in one crop (beets) and the average concentration was 2.5 nanograms of PFOA / gram of beet. The concentrations of GenX and PFOA reported in this report in crops from vegetable gardens in the vicinity of the DuPont / Chemours company provide answers to the first question.

The concentrations measured in 22 samples from the location with the highest concentrations were used as a 'worst case' situation to calculate the intake of GenX and PFOA. The intake calculation is based on a minimum and a maximum scenario. In the minimal scenario the lowest measured concentration per (category) crop is used and in the maximum scenario the highest measured content per (category) crop is used. In the maximum scenario, the TDI's fill for GenX was 36% for the estimate of the average intake and 100% for the estimate of the high intake. The filling of the TDI for PFOA varied in the maximum scenario of 34% for the estimation of the average intake to 96% for the estimation of the high intake. The answer to question 2 reads as follows: No, the permissible daily intake via food from GenX and PFOA is not exceeded by consumption of vegetable crops in a usual consumption pattern.

The actual intake of GenX and PFOA by eating vegetables from the kitchen gardens is probably lower than the calculated maximum intake by, among other things, the following factors:

- the intake calculation is based on consumption of crops from the vegetable garden with the highest concentrations of GenX and PFOA;

- the intake calculation is based on the assumption that only crops from the garden are eaten;

- In a number of cases, the intake calculation is based on concentrations of GenX and PFOA in unwashed and / or unpeeled vegetables.

Like every research, this research also contains uncertainties. Where possible, these uncertainties have been quantified in this study. The intake results are influenced by uncertainties in the concentration data of GenX and PFOA, the consumption data, the link between the measured and consumed products and the model used for the calculation of the intake. The main source of uncertainty in intake is the measured concentrations of GenX and PFOA. This is mainly due to the limited number of samples in combination with a high percentage of samples with a content below the detection limit and the spread in the levels per crop. The derivation of the TDI of GenX and PFOA is based on aspects that each have their own uncertainty. An uncertainty analysis shows that the largest contribution to the uncertainty in the TDI of both GenX and PFOA is provided by differences between test animals and humans in the degree of accumulation in the body and the derivation of the dose where no negative health effects have been observed in the test animal.. In addition, late the uncertainty analysis shows that the sum of the quantifiable uncertainties in the TDIs is higher than the uncertainties in the intake

Finally, the exposure was through vegetable crops and is not the only source of exposure to GenX and PFOA. Drinking water and air are also sources of exposure. Therefore, an estimate has been made of the contribution of the sources of drinking water, air and vegetable crops to the total exposure by expressing each contribution separately as a percentage of the TDI. The total contribution of drinking water, air and vegetable crops to the TDI varied from approximately 57% to 124% in the maximum scenario for GenX and from 46 to 108% for PFOA.
In view of the low contribution of drinking water and the inevitable contribution of air to exposure, the RIVM recommends consuming in a modest way (not too often or too much) vegetable crops that are grown within a radius of 1 kilometer from the farm. Outside this area, concentrations are so low that the crops can be safely eaten, also in combination with other exposure sources.

1 Introduction

1.1 Background

1.1.1 Reason

The Vrije Universiteit Amsterdam (VU) conducted an exploratory study in 2017 into the GenX and PFOA levels in grass and leaves in the vicinity of the DuPont / Chemours company (Brandsma et al., 2017). The highest concentrations in leaves and grass were found at a distance of 50 m from the company (Brandsma et al., 2017).
From 200 to 3000 m, the GenX concentrations ranged from 1.0 - 12.8 ng / g and for PFOA from 0.4 - 19.8 ng / g.

As a result of this research, uncertainty arose about the safety of eating fruit and vegetables from vegetable gardens in this area. On the basis of the VU's results, the RIVM was unable to make a statement about the quantities of GenX and PFOA present in or on edible crops. That is why the Municipality of Dordrecht, for the cooperating authorities, has asked the RIVM to carry out research into vegetable gardens near the DuPont / Chemours factory.

1.1.2 Client

The client for this research is the Municipality of Dordrecht, for the cooperating authorities (Sliedrecht, Papendrecht, Province of Zuid-Holland and Rijk) through the board of B & W.

1.2 Investigated substances: GenX and PFOA

GenX is not strictly a substance, but a technology that is used in the production of fluorinated polymers, such as polytetrafluoroethylene (PTFE, brand name Teflon). GenX technology uses the substance 2,3,3,3-tetrafluoro-2- (heptafluoropropoxy) -propanoic acid (FRD-903), which after reaction with ammonium hydroxide (ammonia) is converted to its ammonium salt: ammonium 2, 3,3,3-tetrafluoro-2- (heptafluoropropoxy) -propanoate (FRD-902). This distinction between the acid and the (ammonium) salt is less relevant to the harmfulness (toxicity) because the effects in the body are caused by the anion in both substances (2,3,3,3, - tetrafluoro-2- (heptafluoropropoxy)) -propanoate). In this report, GenX is therefore the anion of FRD-902 or FRD-903.

PFOA is the abbreviation for perfluorooctanoic acid (from the English perfluorooctanoic acid) and is also an auxiliary in the preparation of teflon. Due to the presence of eight carbon atoms, the less specific abbreviation C8 is also used.

1.3 Purpose and research question

Target

The aim of this research is to find out if people can safely eat the vegetables from a vegetable garden in the vicinity of DuPont / Chemours with regard to the GenX and PFOA concentrations in the vegetables.

Questions

Two questions have been formulated for the kitchen garden research into GenX and PFOA:

1. What are the concentrations of GenX and PFOA in selected crops from vegetable gardens in the vicinity of DuPont / Chemours?
2. Is the allowable daily intake via food from GenX and PFOA exceeded by consumption of vegetable crops in a usual consumption pattern? (Found concentrations of GenX and PFOA in crops are compared with the health standards for lifelong intake via food).

1.4 Study design

1.4.1 Phasing of the research

Different variables may influence the levels of GenX and PFOA in vegetables: type of vegetable, location relative to the plant (distance, wind direction), GenX and PFOA concentration in soil and GenX and PFOA concentration in water used for watering. These variables are included in the study in phases. In the first phase, the GenX and PFOA concentrations are determined in selected crops from different vegetable gardens. Using the concentrations of GenX and PFOA, the daily dietary intake is determined and compared with the allowable daily intake of GenX and PFOA. In the second phase, soil and water samples are analyzed if the crop research shows that the concentrations of GenX and PFOA are so increased that additional analyzes in soil and water are needed to examine the origin (via soil or water). The results of the first phase of the research (crop research) are described in this report.

1.4.2 Intended study design for the first phase

The proposed research has a limited design (limited in number of locations, crops to be examined and time interval). In consultation with the municipalities, a number of locations will be selected with a different distance and wind direction from the DuPont / Chemours plant. The choice of sample locations creates an indicative picture for (vegetable) gardens in the Dordrecht, Sliedrecht and Papendrecht area. A reference location is included in the study.

From the vegetable crops a selection is made of crops that make up a relatively large share in the kitchen garden menu (a leafy vegetable, a bulbous vegetable and a fruit vegetable per location).
Fruit crops (from the vegetable garden as well as in the wild, for example blackberries) are not (everywhere) included in the analyzes, because fruit consumption is expected to be a smaller share of the consumption of vegetable crops. At the foreseen start of sampling (end of August) the harvest season is largely over; sampling of early crops is no longer possible.

If possible, one representative vegetable is taken at each selected location of each category of vegetable (leaf, tuber and fruit vegetable). As an example: courgettes, cucumbers or tomatoes are used for the vegetable fruit category. Five plants (or more) of each representative vegetable, if present, are sampled. A mixed sample will be made in the analysis laboratory (so five plants from one representative vegetable form one sample). If sufficient plants are available per vegetable, they will be split into a treated (wash or peel) and an untreated sub-sample prior to mixing.

At one of the above locations, with possibly the highest load by the factory, two representative vegetables and two fruit plants are sampled per category of vegetables. This is done to gain some insight into the variation within a category of vegetables.

1.5 Implementation

At the end of August 2017 samples of vegetables were taken at 10 locations near the Dupont / Chemours factory. This concerned three locations in Dordrecht, three in Papendrecht and four in Sliedrecht. The locations have been designated by the relevant municipalities. In addition, a reference site in Bilthoven has been sampled.

It turned out to be possible to take samples at all locations of the three selected categories of vegetables (leaf, tuber and fruit vegetables). More crops were sampled at one of the locations, namely six vegetables (two per category) and two fruit crops (apple and pear). Moreover, it turned out to be possible to take several plants from almost every vegetable and split them into an untreated and treated sub-sample. Only the only monster pumpkin is not split. To prevent confusion, these split sub-samples are referred to as 'samples' later in the report.

In this way, of 11 locations (including the reference location)
81 samples obtained and analyzed by the research institute RIKILT of Wageningen University & Research (WUR) for the presence of GenX and PFOA.

The locations around the Dupont / Chemours factory are numbered 1 to 10 in Figure 1 and, for privacy reasons, are not specified.

1.6 Classification of the report

Following this introductory chapter, Chapter 2 describes the methodology for risk assessment. In chapter 3 the results are described and in chapter 4 a discussion is delivered and the conclusions are drawn.

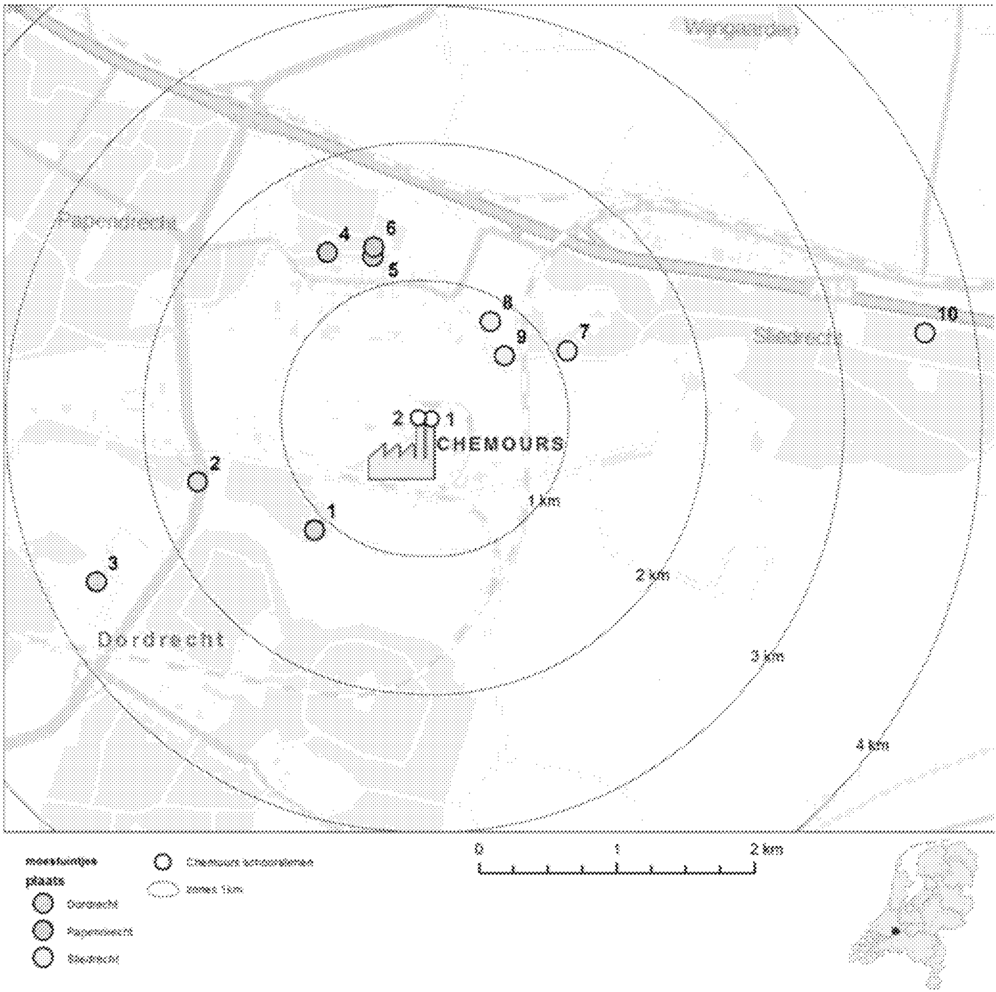

Figure 1. Representation of the 10 locations of vegetable gardens around the DuPont / Chemours factory where samples of crops were taken in August 2017.

2 Method of risk assessment

2.1 Introduction

To carry out a risk assessment of GenX and PFOA present in vegetables and fruit in vegetable gardens, the intake of these substances was compared with the allowable daily intake (TDI) of GenX and PFOA. For the calculation of intake, the measured concentrations in the sampled vegetable crops were combined with the consumption quantities of vegetables and fruit from two food consumption surveys conducted in the Netherlands. Where possible, the uncertainties in the data used were quantified in the interpretation of the results.

2.2 Concentrations

The samples were analyzed according to the method described in RIKILT SOP-A-1114. During the sample workup, the internal standards (13C-PFOA and 13C-GenX) were added to the samples prior to an extraction and solid phase extraction (WAX-SPE) clean-up. The sample extracts were then analyzed using LC-MSMS (LC - Shimazdu Nexera X2 LC-30AD UHPLC; MS - AB Sciex Qtrap 5500 triple quadrupole mass).

Table 1 shows the detection limit (LOD) and quantification limit (LOQ) for the analysis of GenX and PFOA in the examined fruit and vegetables.

Table 1. Detection limit (LOD) and quantification limit (LOQ) for GenX and PFOA in the crops tested (in ng / g wet weight).

Dust	Crop	LOD	LOQ
GenX	All	0,5	1,0
PFOA	PFOA Potatoes	0,4 0,1	1,0

Three types of measurement results could be distinguished:

1. The concentration was above the LOQ and therefore the concentration is expressed in a number,

2. The concentration was below the LOD and then that sample is indicated as smaller than the detection limit (<LOD),

3. The measured concentration was below the LOQ but above the LOD and this sample is indicated with a plus (+).

Two measurements were made per sample, so-called duplicate measurements. If the concentration in both measurements was above the LOQ, an average concentration was calculated (see section 2.5.2).

2.3 Statistical analysis

Based on the chemical analyzes of the vegetables sampled at all locations (including reference), a statistical analysis was performed on the data. Due to the very limited number of samples of fruit (taken at one location) the measurement results of the fruit were not included in the statistical analysis.

Based on a statistical analysis, an attempt was made to get an answer to the following questions:

1. Are there clusters to be discovered in the vegetable garden locations on the basis of the supplied data on the content of GenX or PFOA in the various vegetable samples?
2. Is there a difference in the level of GenX or PFOA content between crops and unwashed products?
3. Is there a difference in the level of GenX or PFOA content between the different vegetable varieties?

2.4 Food Consumptions

For the calculation of the intake of GenX and PFOA via vegetable crops, the consumption data from two Dutch food consumption surveys (VCPs) were used: the survey among children aged 2 to 6 years (Ocké et al., 2008) and the survey among persons aged 7 up to and including 69 (Van Rossum et al., 2011). In these polls individuals have indicated on two days what they have consumed and drunk, including the quantities consumed and drunk.

2.5 Taking GenX and PFOA

2.5.1 Coupling measured concentrations of consumed products

For the calculation of the intake of GenX and PFOA on the basis of the measured concentrations in vegetable crops, a link was made with the consumed products from both VCPs. The vegetables that have been measured are divided into three categories: leaf, tuber and fruit vegetables. The fruit measured involved apples and pears and these are classified in the category pome fruits. It is known that more vegetables are grown in the relevant vegetable gardens that were not present during the sampling.

That is why two extra vegetable categories have been added: cabbage and leguminous vegetables. For intake via fruit only the consumption of apple and pear was included in the analysis.

For the classification of the vegetables consumed in the various categories, see Appendix 1.

2.5.2 Used concentrations

The number of sampled crops per category was limited (n = 3 for various vegetable categories and n = 2 for fruit). The calculation of the intake is therefore based on a minimal and maximum scenario. In the minimal scenario, the lowest measured concentration per crop or category is used for the intake calculation and in the maximum scenario the highest measured content per crop or crop category.

As described in section 2.2, the measured concentrations of GenX and PFOA in the vegetable crops were not only reported as a positive concentration (above the LOQ), but also as a value between the LOQ and LOD (+), or as a value under the LOD . Per a duplicate measurement was performed. See Table 2 for the allocation of a minimum and maximum concentration per duplometry. In the case of two positive concentrations (above the LOQ), the geometric mean is calculated.

After allocation of the concentrations to the measured replicates, the minimum and maximum concentration per crop, regardless of preparation or category, is selected and linked to the relevant consumption quantities for the calculation of the intake in accordance with a minimal and maximum scenario.

Table 2. Award of GenX and PFOA concentrations per duplometry

Result of the duplometry	Minimale concentratie	Maximale concentratie
2 x < LOD	½ x LOD	LOD
1 x < LOD and 1 x < LOQ[1]	LOD	LOQ
2 x < LOQ	LOD	LOQ
1 x < LOQ and number (=1 x > LOQ)	LOQ	Number
2 x numberl (= 2 x > LOQ)	Geometric Mean[2]	

LOD = detection limit; LOQ = quantification limit 1 <LOQ = value between LOD and LOQ 2 Calculated concentration does not differ between the minimum and maximum scenario

2.5.3 Used model for calculation of intake

GenX and PFOA may be detrimental to health during long-term exposure. For the calculation of this long-term intake, the Observed Individual Mean (OIM) model, as implemented in the Monte Carlo Risk Assessment (MCRA) software version 8.2 (De Boer et al., 2016), was used.

With this model, the quantities of products consumed by individuals per day are multiplied by the measured GenX and PFOA concentration per product. The intake per product is then summed per person over the products, resulting in the intake per person per day. This intake is then divided by the body weight (lg) of the individual in question. Because intake is important over a longer period, the average intake per person is then calculated over the two days in the VCPs (section 2.4). This resulted in a distribution of individual average daily intake. The intake is then quantified as the mean, median (50th percentile, P50) and high (95th percentile, P95) intake for the entire Dutch population aged 2 to 69 years.

This calculation was performed for GenX and PFOA with both the minimum and maximum concentrations per crop or category (section 2.5.2).

2.6 Toxicity of GenX and PFOA

2.6.1 General

For the risk assessment, the calculated intake was compared with the allowable daily intake (TDI) for GenX and PFOA as derived by the RIVM (Janssen, 2017; Zeilmaker et al., 2016). The TDIs for GenX and PFOA are 21.0 and 12.5 ng / kg bw / day respectively.

Two RIVM reports indicate that in the (recent) past several reputable international bodies have derived various health-based limit values for PFOA (Zeilmaker et al., 2016; Oomen & Herremans, 2017). It is expected that the European Food Safety Authority (EFSA) will issue a (new) scientific opinion in 2018 on the risk assessment of PFOA in food.

In sections 2.6.2 and 2.6.3, a brief description will be given of the basis on which the RIVM derived the TDIs for GenX and PFOA.

2.6.2 GenX

In the report by Beekman et al. (2016) a limit value for the inhalation of FRD-903 for the general population is derived. Since all available toxicity studies have been performed with the ammonium salt (FRD-902) and not with the acid (FRD-903), the limit value of FRD-903 is based on the FRD-902 data. It is justified to use FRD-902 data for

FRD-903, because the effects in the body on both substances are caused by the anion (see section 1.2).

This limit value for inhalation is based on the dose where no adverse health effects have been observed in the test animal, a so-called No Observed Adverse Effect Level (NOAEL) of

0.1 mg / kg bw / day. This NOAEL is derived from an oral, chronic rat study with the substance FRD-902 and is based on an immunotoxic effect (effect on albumin / globulin ratio). Based on this NOAEL in rats and some uncertainties, a preliminary TDI of 21 ng / kg bw / day was derived for GenX. The TDI is qualified as 'provisional' because of the uncertainty about GenX kinetics in humans. For details on the derivation of this TDI, see Appendix 2.

2.6.3 PFOA

In 2016, RIVM derived a health-based limit value for PFOA, taking into account the accumulation of this substance in the human body through prolonged exposure (Zeilmaker et al., 2016). Studies with laboratory animals have shown that liver toxicity is the most sensitive effect of PFOA. Based on a so-called NOAEL in rats and some uncertainty factors, a TDI of 12.5 ng / kg bw / day was derived for PFOA (Zeilmaker et al., 2016).

The report by Oomen and Herremans provides an overview of a number of health-based limit values for the long-term exposure to PFOA (Oomen & Herremans, 2017). These values range from 2 to 1500 ng / kg bw / day. The highest value comes from an EFSA opinion of 2008 and is expected to be revised downwards due to advancing scientific insight. The lowest value comes from the New Jersey Drinking Water Quality Institute (NJDWQI) (NJDWQI, 2016).

2.7 Risk assessment

2.7.1 General

Both the intake calculation and the risk assessment are based on daily consumption of vegetable crops throughout life, a so-called chronic intake of both GenX and PFOA. For the risk assessment, it is customary to compare the intake against the TDI. A marginal note is that the levels used in fruit and vegetables will vary over time. These variations may depend on general factors such as weather conditions and seasonal variation, but also on substance-specific factors such as the fact that the use of PFOA in the DuPont / Chemours plant was stopped in 2012 and the use of GenX started in 2012. It is therefore good to realize that the specific crop / product concentrations, and thus the calculated intakes, are a snapshot.

2.7.2 Comparison of exposure with the TDI

In the first instance, the calculated exposure of GenX and PFOA (conservative value) was compared with the TDI. For GenX, the TDI of 21.0 ng / kg bw / day was used and for PFOA the TDI was 12.5 ng / kg bw / day. This is the usual method.

A comment on the use of the TDI for PFOA derived by the RIVM is, as indicated in section 2.6.1, that EFSA may derive a different TDI in the near future. The comment at the TDI of GenX, also derived from the RIVM, is the (great) uncertainty about GenX kinetics in humans (section 2.6.2).

2.7.3 Refinement of the risk assessment with APROBA-Plus

The risk assessment whereby the intake is compared with the TDI has been refined with the APROBA-Plus method. With this method, the uncertainties in intake (exposure) and toxicity are quantitatively evaluated (Bokkers et al., 2017). This method is an extension of a methodology developed by the World Health Organization (WHO) (WHO-IPCS, 2014) and results in a better understanding of the risks of exposure to substances. The APROBA-Plus method was also used in this study to gain insight into the contribution of the various sources of uncertainty to the total uncertainty in the risk assessments of PFOA and GenX.

Below we briefly describe which uncertainties for intake and toxicity are quantified in the APROBA-Plus method. These are the same sources of uncertainty that are also taken into account in the usual method (see section 2.7.1) (but less transparent and complete).

Intake

As discussed in section 2.5.1, for the intake calculation the levels of GenX and PFOA in crops are linked to the consumed quantities of these crops on the basis of consumption data from the general Dutch population. The uncertainties in the intake calculation are therefore mainly related to the uncertainty in the concentrations (including the concentrations in the non-measured crops), the uncertainty in the applicability of the consumption quantities of fruit and vegetables for vegetable users and the link between the measured and consumed Products. The uncertainties in the measured concentrations are quantified by the use of a minimal and maximum scenario and thus included in the APROBA-Plus method. The other sources of uncertainty can only be assessed qualitatively and are not included in the APROBA-Plus method.

Toxicity

In the APROBA-Plus method, the uncertainties in the derivation of the TDI of GenX and PFOA have been quantified. This mainly concerns the uncertainties concerning the NOAEL used and the extrapolation steps used to the human target population (such as differences in kinetics and dynamics within a species (intraspecies) and between species (interspecies)).

3 Results

3.1 Measured concentrations of GenX and PFOA in fruit and vegetables

Appendix 3 shows the results of the chemical analyzes of GenX and PFOA in vegetables and apple and pear from the various locations. The authors only knew that the reference location was the location with code G4LOC1 (see also section 3.3). Table 3 gives an overview of the total number of analyzed samples and the number of samples with a content under the LOD, between the LOD and LOQ and higher than the LOQ for the reference location and the locations around the DuPont / Chemours plant.

Table 3. Number of analyzed samples for the reference site and the locations around the DuPont / Chemours plant and the number of samples with an analyzed content below the detection limit (LOD), between the LOD and the quantification limit (LOQ) and above the LOQ1

Location	Number of Samples			
	Total	< LOD[2]	Between LOD en LOQ[3]	≥ LOQ[4]
Reference	7	GenX: 7 PFOA: 7	GenX: 0 PFOA: 0	GenX: 0 PFOA: 0
Around Factory	74	GenX: 45 PFOA: 44	GenX: 19 PFOA: 27	GenX: 10 PFOA: 3

1 For the concentrations associated with the LOD and LOQ, see Table 1.
2 Measurement measurements were both <LOD
3 Plot measurements were both <LOQ, possibly one lower than LOD
4 At least one of the duplicate measurements had a level> LOQ

Based on the results, the following conclusions can be drawn:

1. Only Genotope and / or PFOA were detected at the reference site on either of the analyzed crops (Table 3). GenX and / or PFOA has been demonstrated at all other locations.

2. No GenX and no PFOA were detected in approximately 60% of the 74 samples taken at the sites around the DuPont / Chemours plant (Table 3).

3. GenX could be quantified in 14% of the samples taken at the sites around the DuPont / Chemours plant. PFOA could be quantified in 4% of these samples.

4. GenX has been quantified in five crops (endive, beet, celery, lettuce, and tomatoes) and PFOA has been quantified in one crop (beets).

5. The highest GenX concentration was 5.9 ng / g in unwashed lettuce, and the highest PFOA concentration was 2.8 ng / g in unwashed and unpeeled beet. Due to the duplicate measurement, the

highest average for GenX was 5.4 ng / g in unwashed lettuce and for PFOA 2.5 ng / g in unwashed and unpeeled beet.

6. In all locations where GenX and PFOA was detected, crops were also detected in which no GenX and PFOA were detected.

7. No GenX has been detected at four of the 10 sites around the DuPont / Chemours plant and no PFOA at two of the 10 sites.

3.2 Statistical analysis

The results of the statistical analysis are described on the basis of the questions as stated in section 2.3. The detailed statistical analysis is included in Appendix 4.

3.2.1 Clusters of vegetable garden locations

The statistical analysis showed that on the basis of the concentrations in the vegetables for both GenX and PFOA, two different clusters (groups) of vegetable garden locations could be identified. For GenX, the locations G3LOC2 and G3LOC4 were not significantly different. As a cluster of two locations, however, they were different from the other locations. For PFOA, the locations G2LOC1, G2LOC3, G3LOC2 and G3LOC4 formed one cluster. The concentrations of PFOA in vegetables from these gardens were significantly higher than the concentrations in the other gardens.

3.2.2 Influence of washing on the concentrations of GenX or PFOA

For the vegetable samples containing GenX there was sufficient evidence that washing a vegetable led to a lower concentration of GenX in vegetables. When analyzing the influence of washing on the samples containing PFOA, it explained how the so-called "+" observations were taken (section 2.2). When these observations were assigned a level of 0.5 ng / gram (= ½ LOQ), there was a difference between crops and unwashed vegetables.

If these positive observations were assigned a level of 1.0 ng / gram (= LOQ), the difference found between crops and unwashed vegetables could also come by chance.

3.2.3 Differences in concentrations of GenX or PFOA between categories of vegetables

For both GenX and PFOA there was a significant difference in concentrations between the different categories. M.a.w. the concentrations of GenX or PFOA in the three categories studied (leaf, tuber and fruit vegetables) were significantly different.

3.2.4 Consequences of the statistical analysis for the intake calculation The results of the statistical analysis have the following consequences for the intake calculation:

- For both GenX and PFOA, G3LOC4 belonged to a cluster that distinguished itself from the rest. Since for both substances (GenX and PFOA) the highest concentrations were found on G3LOC4, these concentrations were used as a 'worst case' situation to calculate the intake of GenX and PFOA.

• The influence of washing of the examined samples was not unambiguous for GenX and PFOA. For the intake calculation, therefore, a possible distinction (for GenX and for PFOA not) in crops / unwashed vegetables is not included.

• The levels of GenX or PFOA in the three categories of vegetables studied are different and that distinction is also included in the intake calculation.

3.3 Link between code and numbering location

To ensure the independence of the research, the link between the code (see Appendix 3) and the numbering of the locations near DuPont / Chemours (Figure 1) not known to the authors. Only the code of the reference location was known. At the request of the client, after the delivery of the draft version of the report, the above link has been announced.

Table 4 shows the link between code and numbering of the location. Table 4. Numbering, coding and position of the locations where samples of crops were taken.

Number	Coding	Location	Position relative to factory	Distance (km)
1	G1LOC1	Dordrecht	SW	1-2
2	G1LOC2	Dordrecht	SW	1-2
3	G1LOC3	Dordrecht	SW	2-3
4	G2LOC1	Papendrecht	NW	1-2
5	G2LOC2	Papendrecht	NW	1-2
6	G2LOC3	Papendrecht	NW	1-2
7	G3LOC1	Sliedrecht	NE	1-2
8	G3LOC4	Sliedrecht	NE	< 1
9	G3LOC2	Sliedrecht	NE	< 1
10	G3LOC3	Sliedrecht	E	3-4
11	G4LOC1	Bilthoven	n.v.t.	> 50

1 O: east; NO: north-east; NW: north-west; SW: south-west

3.4 Taking GenX and PFOA

3.4.1 Concentrations

Because the measured levels differed for the three categories studied, the intake was calculated using the (average of two measurements) concentrations of GenX and PFOA per measured category. The concentrations for a measured representative of a category (eg endive or lettuce for leaf vegetables) are directly linked to their consumption. For the other products within a category (Appendix 1) a value is assigned based on the contents of these representatives. For two categories, cabbage and leg vegetables, no representatives were measured. Vegetables belonging to these categories are linked to the minimum and maximum measured levels across all representatives, regardless of the category to which they belong. Table 5 gives an overview of the used GenX and PFOA levels in the calculation for both scenarios on the basis of the measured concentrations in G3LOC4. These concentrations are derived from the measured concentrations (Appendix 3) as described in

section 2.5.2.

The effect of preparation is not included (section 3.2.4). However, an exception has been made for potato. The consumption of this bulbous vegetable has been reported in the VCPs as boiled potato without peel ("potato z peel cooked"). The consumption of potato is therefore linked to a minimum and maximum GenX and PFOA content for peeled carrot and beet, both representatives for tuber vegetables (Table 5).

Table 5. Minimum and maximum concentrations of GenX and PFOA as used in the intake calculation

		Concentratie (mg/kg)			
		GenX		PFOA	
Category	Crop[1]	Minimaal	Maximaal	Minimaal	Maximaal
Leafy Vegetables	Endive	0,5	1	0,05	1
	Lettuce	1,76	5,38	0,05	1
	Other leafy vegetables	0,5	5,38	0,05	1
Tuber Vegetables	Beet	0,5	2,46	1,44	2,48
	Carrot	0,25	1	0,1	1
	Peeled Potatod[2]	0,25	1	0,1	1,71
	Other tubers	0,25	2,46	0,1	2,48
Fruit Vegetables	Bell Peppers	0,25	0,5	0,05	0,1
	Tomatoes	2,86	3,32	0,1	1
	Other	0,25	3,32	0,05	1
Stone Fruit	Apple	0,5	1	0,05	0,1
	Pear	0,25	1	0,05	0,1
Cabbage	All	0,25	5,38	0,05	2,48
Legumes	All	0,25	5,38	0,05	2,48

1 For crops belonging to 'other' and 'all', see Appendix 1

2 Consumption of potatoes is linked to the concentrations of GenX and PFOA measured in peeled beet and carrot

3.4.2 Food consumption

Data from two VCPs were used for food consumption data (section 2.4). For a number of vegetables and fruits that contribute substantially to the intake of GenX and PFOA

(section 3.4.3) is the average consumption, the percentage of days within the VCPs on which the consumption of the product is reported and

Table 6. Average consumption per product, the lover's portion and the percentage of consumption days on which the consumption of the product has been reported

Product	Consumption (g per day)		Percentage Consumption days (%)[2]
	Average	Lovers Portion[1]	
Cooked potato peel	60	310	40
Apple Peel	90	150	16
Apple Husk	130	260	14
Cauliflower raw/cooked	130	240	4
Cooked broccoli	110	240	3
Raw tomatoe	60	130	6
Raw lettuce crop	40	100	3
Raw cucumber peel	60	140	6
Cooked frozen spinach	100	240	2

1 Lover's portion is the 95th percentile of the consumption quantity on the days on which the consumption of the food has been reported

2 The sum of the number of consumption days in both VCPs is 101963

the enthusiastic portion shown in Table 6. The fancier portion is calculated as the 95th percentile of the consumption of the days on which the consumption of the relevant product is reported. The lover's portion for potato is 310 g (Table 6). This means that when respondents in the VCPs indicated that they had eaten potato in one day, 95% of these respondents

310 g has eaten; 5% of the respondents reported the consumption of a larger amount.

For the calculation of the intake of GenX and PFOA the reported consumption quantities per individual have been used (see section 2.5.3 for the method). The intake is not calculated on the basis of the average consumption or enthusiast portions as shown in Table 6.

3.4.3 Inception calculations

GenX

The average and median intake of GenX for the population of

2 to 69 years varied between 1.8 and 7.6 ng / kg bw / day and 1.2 and 5.6 ng / kg bw / day respectively based on the measured minimum and maximum concentrations at location G3LOC4 (Table 7). For people with a high consumption of vegetables, apple and / or pear, the intake of GenX varied between 5.2 and 21 ng / kg bw / day.

Table 7. Taking GenX and PFOA per scenario in the population aged 2 to 69 years

Intake	Intake (ng/kg lg/dag)			
	GenX		PFOA	
	Minimaal	Maximaal	Minimaal	Maximaal
Average	1,8	7,6	0,3	4,3
P50	1,2	5,6	0,2	3,0
P95	5,2	21	0,8	12

In Figure 2 the contributions of the categories to the total intake distribution of GenX are shown for the maximum scenario. In this scenario, cabbage vegetables contributed the most to the intake by 21%, followed by leaf, tuber and fruit vegetables (18-19%).

Leguminous vegetables with 11% contributed least to the exposure.

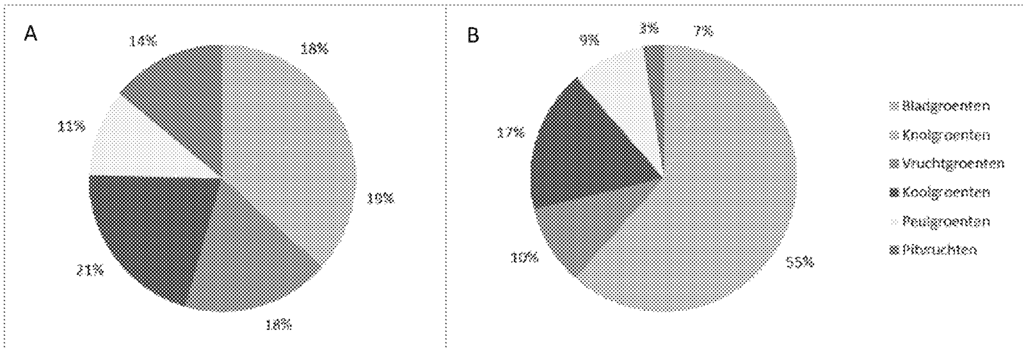

Figure 2. Contribution (%) of the different categories of fruit and vegetables to the total intake distribution of GenX (A) and PFOA (B) in the Dutch population from 2 to 69 years.

Within the cabbage vegetables, the largest contribution was made by cauliflower (32%) and broccoli (25%). These contributions from cabbage vegetables were driven by the consumption data, because the same GenX concentration was used for all cabbage consumption in the category of cabbage vegetables (Table 5). The same GenX concentration was used for leguminous vegetables as for cabbage vegetables (Table 5).

Leguminous vegetables, however, contributed less to exposure due to lower consumption quantities.

The product that contributed most to the overall intake of GenX was potato (15%), followed by apple (12%), lettuce and tomatoes (8% each), cucumber and cauliflower (7% each), and broccoli and spinach (5%). Other products contributed less than 5% to the intake of GenX in the maximum scenario.

PFOA

For PFOA, the intakes were lower than for GenX (Table 7) due to lower measured concentrations (Table 5). The mean, median and high (P95) intake varied between 0.3 and 4.3, 0.2 and 3.0, respectively

and 0.8 and 12 ng / kg bw / day.

Tuberous plants contributed most to the exposure to PFOA in the maximum scenario: 55% (Figure 2). This was followed by cabbage and fruit vegetables with a contribution of 17% and 10% respectively. The contribution of bulbous vegetables to the intake of PFOA in the maximum scenario was determined almost entirely (90%) by potato through a combination of a high consumption frequency (Table 6) and a high PFOA content (Table 5). Cabbage vegetables contributed, as with GenX, through the consumption of cauliflower and broccoli.

The potato product contributed almost 50% to the total intake of PFOA in the maximum scenario, followed by green beans (7%) and cauliflower (6%). Other products contributed less than 5%.

3.5 Uncertainty analysis

3.5.1 Intake

The calculated intakes are influenced by uncertainties in the concentration data of GenX and PFOA, the consumption data from the VCPs and the link between the measured and consumed products. The model used also gives an uncertainty in the calculated intakes.

Concentrations

Various uncertainties play a role in this research. The main uncertainty was the measured concentrations of GenX and PFOA. First, the concentration measurements have to do with a LOD and / or LOQ, i.e. a limit in the measuring method below which the concentration can no longer be detected or quantified. In case the concentration is lower than the LOQ but larger than the LOD, a concentration range is known. When the concentration is lower than the LOD it is not known how much lower.

In addition, the uncertainty in the concentrations is determined by the limited number of samples in combination with a high percentage of samples with a level below the LOQ and LOD and the distribution in the levels per crop (category). To include this uncertainty, a minimum and maximum concentration has been calculated (Table 5), resulting in two intake scenarios. These reflect the bandwidth of the intake of GenX and PFOA on the basis of the measured concentrations. The intake of GenX differed for the high (P95) intake by a factor of 4 between the minimum and maximum scenario, whereas for PFOA it was a factor of 15. Because of the use of maximum concentrations combined with the choice of the concentrations of the site with the highest concentrations (G3LOC4), the intake calculated with the maximum concentrations can be considered conservative on the basis of the available data.

GenX and PFOA can be detrimental to long-term health. The measurements provide a picture of the concentrations at the time of sampling. The use of PFOA was stopped in 2012 and that of GenX started in 2012. The concentrations of both substances in crops have therefore varied in the past and will also change in the future. The calculated intake forms a snapshot and can only give a limited impression of the intake over a longer period.

Consumption data

The uncertainty regarding the consumption data is mainly due to the representativeness of the consumption data of vegetables, apple and pear from the average Dutch population for vegetable gardeners. The consumption data comes from a representative sample of the Dutch population.

Vegetable gardeners, however, may consume more of the fruit and vegetables they have grown than the average Dutch population. A study from 2007 shows that the average consumption of home-grown potatoes by vegetable gardeners (and their families) is a factor 1.1 higher than for the average Dutch person. For the other vegetables, this is a factor of 1.2 for babies and non-school children and a factor of 1.7 for school-age children and adults (Swartjes et al., 2017). These factors are based on a dietary study of 154 households with vegetable gardens from 1988 and the Dutch Food Consumption Survey of 1997/1998. As a result, the factors are probably no longer accurate, but do show that it is very likely that vegetable gardeners consume the vegetable crops they grow in larger quantities than the average Dutch person. However, by calculating the high intake (P95) of GenX and PFOA, higher consumption than the average consumption of vegetable crops was included in this study.

Link measured and consumed products

For the calculation of the intake of GenX and PFOA, a link has been made between the measured and consumed products. Different categories were defined for this because there were more consumed vegetables than were analyzed. This link assumes that the concentrations of GenX and PFOA in the measured crop types were representative of the levels in all vegetables belonging to such a category. The measured levels in beet and carrot are also attributed, for example, to the consumption of potato, swedes, peas with carrots, snow peelings, celeriac and radish (Appendix 1). This choice was made because vegetables other than the vegetables that have been sampled can also be grown in a vegetable garden and to minimize underestimation of the intake due to this uncertainty.

No measurement data were available for two vegetable categories: cabbage and legumes. The consumption quantities of vegetables belonging to these categories, such as cauliflower, broccoli and green beans, are therefore linked to a minimum and maximum measured concentration of GenX and PFOA in all analyzed vegetables. This meant, for example, for the maximum scenario that the consumption of all vegetables belonging to cabbage and legumes is linked to the maximum GenX content (found in unwashed lettuce) and the maximum PFOA content (found in unwashed and unpeeled beet) (Table 5).

By linking the analyzed levels of GenX and PFOA to all reported consumptions of vegetables, apples and pears in the VCPs, it is assumed that vegetable gardeners do not consume purchased vegetables, apples and pears that do not contain GenX and PFOA. And that they do this over a very long period. Most vegetable gardeners, however, will also consume commercially grown products, eg during the winter period. This assumption may therefore have resulted in an overestimation of the intake of GenX and PFOA by vegetable gardeners.

Effect of preparation

In the intake calculations the effect of washing and peeling on the GenX and PFOA levels was not included, except for potato (section 3.4.1). For the 10 vegetable samples to which GenX could be quantified, there was evidence that washing a vegetable led to a lower concentration of GenX (section 3.2). This did not apply to PFOA, but this conclusion was based on concentrations measured in one crop, namely beets. The concentrations in crop beets were lower than in unwashed beets. By not including a possible effect of washing in the intake calculation, the intake may have been slightly overestimated. However, we estimate that the effect would not have been significant, because potato contributed most to the exposure of both substances (section 3.4.3). For potato, the effect of washing has been included in the analysis.

Used model

The OIM model was used for the calculation of the intake (section 2.5.3). With this model, the average intake over the available days in the VCPs, in our case two, is used as predictor of the long-term intake. Calculations have shown in the past that such a model can well predict the average and median intake in a population. However, these models overestimate the intake in the right tail of the distribution, such as the P95, because the number of days on which people report their food consumption is too limited for this (Boon & van der Voet, 2015). There are models available to better estimate the intake in the long term on the basis of, for example, only two days. However, such a model has not been used here in view of the limited concentration data set.

summarizing

Various sources of uncertainty may have led to underestimating or overestimating the intake of GenX and PFOA by vegetable gardeners. All in all, we estimate that the calculated intakes on the basis of the maximum scenario give a conservative estimate of the actual intake of GenX and PFOA by vegetable gardeners living in the vicinity of DuPont / Chemours. The minimal scenario may underestimate the intake

3.5.2 Toxicity

The underlying aspects used in deriving the TDIs for GenX and PFOA each have their own uncertainties. Both for GenX and for PFOA the standard uncertainty distributions as published by WHO-IPCS (2014) were used for these aspects, except for the scaling of the dose from test animal to human. For PFOA it has been calculated that the translation from the dose of test animal (rat) to humans is approximately a factor of 60, due to the accumulation time that is longer in humans, with an uncertainty factor of 2 (uncertainty range 30-120, see Appendix 5A, Table 5A .2). For GenX this uncertainty is greater (uncertainty range 15-240, see Appendix 5B, Table 5B.2). In both cases, this factor replaces the standard allometric factor for scaling differences in kinetics.

The largest contribution to the uncertainty in the toxicity of GenX was due to differences in kinetics between test animals and humans (25 + 16 = 41%), followed by the uncertainty in the NOAEL (31%) and intraspecies differences (28%). See Appendix 5B for calculations performed with APROBA-Plus.

The largest contribution to the uncertainty in the toxicity of PFOA was caused by the uncertainty in the NOAEL (29%), closely followed by differences in kinetics within an animal species (26%) and the relatively short duration of the laboratory animal experiment (24%).). See Appendix 5A for calculations performed with APROBA-Plus.

3.6 Risk assessment

3.6.1 GenX

Comparison of exposure with the TDI

For the risk assessment, the calculated intake of GenX, based on a consumption pattern of the Dutch population, was compared with the TDI of 21 ng / kg bw / day. All intake estimates (mean, P50 and P95), except for one, were below the TDI in both the minimum and maximum scenarios. One estimate was on the TDI. In order to take account of other sources of exposure, the "filling" of the TDI is often looked at. The filling of the TDI varied from 6% for the median (P50) estimate in the minimal scenario to 100% for the high intake in the maximum scenario.

Refinement of the risk assessment with APROBA-Plus

Figure 3 shows the result of the risk assessment with the APROBA-Plus method. This assessment is based on the same assumptions and data (both in terms of toxicity and exposure), but with a more precise quantification of the uncertainties. APROBA-plus estimates the human dose at which the relevant immunotoxic effect (change in albumin / globulin ratio) would occur in 1% of consumers.

The vertical blue line indicates where the human dose might be, which causes 1% of consumers immunotoxic effects. The horizontal blue line indicates where the intake might lie for the 95th percentile of the population. Note the logarithmic scale of both axes. The ellipse that encompasses both lines thus indicates where the combination of the actual value of human dose and exposure (as just defined) might lie. It can clearly be seen therein that the entire ellipse is in the green (i.e., the "safe") area. Moreover, it is striking that the uncertainty in the human dose (upper and lower limit of the ellipse) is more than a factor of 10 greater than the uncertainty in the intake (left and right border of the ellipse).

The marked point (blue diamond) in this figure shows the combination of the TDI of 21 ng / kg bw / day and the high intake based on the maximum scenario, according to the usual method. This illustrates that the usual method gives a very limited picture of reality and the available knowledge about it.

3.6.2 PFOA

Comparison of exposure with the TDI

For PFOA, all calculated intakes in both scenarios were below the TDI of 12.5 ng / kg bw / day. The filling of the TDI varied from 2% for the average and median estimate of the intake in the minimal scenario to 96% for the estimation of the high intake in the maximum scenario.

Refinement of the risk assessment with APROBA-Plus

Figure 4 shows the result of the calculation with the APROBA-Plus method (see section 3.6.2 for an explanation of the figure). Again, it appears that the entire ellipse is in the green (i.e., the "safe") area. Again, the uncertainty in the dose (upper and lower limit of the ellipse) is greater than the uncertainty in the high intake (left and right border of the ellipse).

The blue diamond in this figure shows here the combination of the TDI of 12.5 ng / kg bw / day and the P95 intake on the basis of the maximum scenario, namely 12 ng / kg bw / day. Again, the figure illustrates that the blue diamond (i.e., the usual method) gives a very limited picture of the reality and the available knowledge about it.

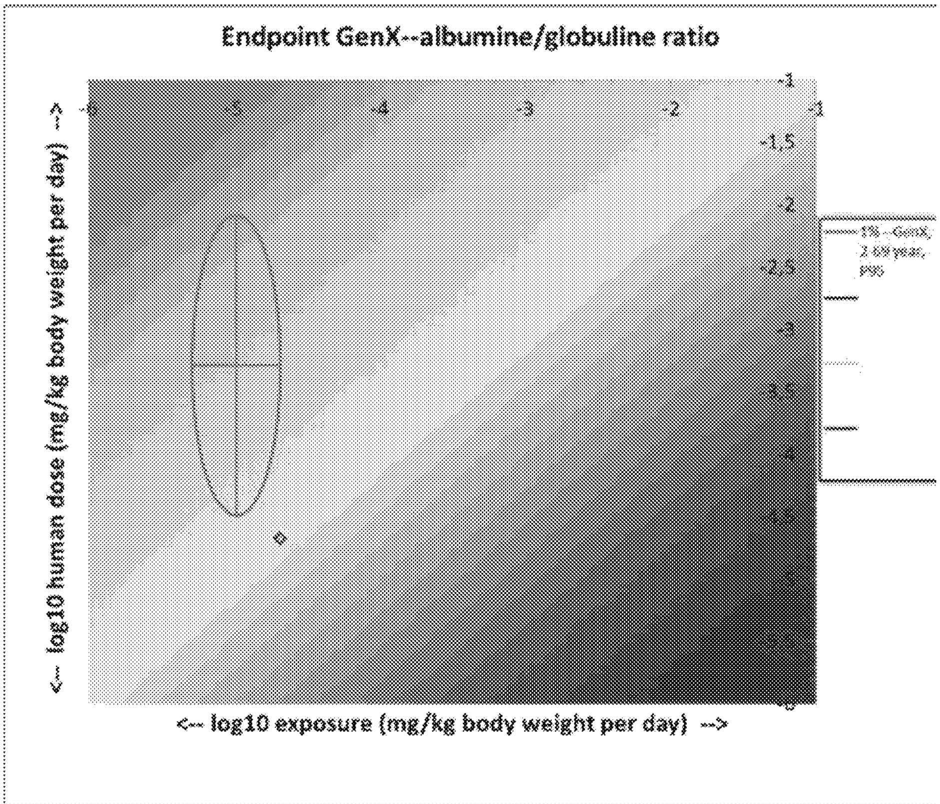

Figure 3. Graphical representation of the uncertainty in the toxicity of GenX ('human dose', the dose that would cause immunotoxicity in 1% of the consumers, represented by the vertical blue line) and the intake ('exposure' to the x- axis, the level of the P95 intake of consumers, represented as the horizontal blue line) of GenX through the consumption of vegetable crops

3.6.3 Conclusions

As explained in section 3.5.1, we estimate that the calculated intake based on the maximum scenario is a conservative estimate of the actual intake of GenX and PFOA by vegetable gardeners in the vicinity of DuPont / Chemours.

In section 3.6.1 and 3.6.2 it is described that in reality the high intake is very likely to be lower than the dose at which 1% of the population could experience harmful effects.

In summary, it can be concluded that, taking into account the uncertainties in intake and toxicity, even the highest fraction of the population that consumes from their own vegetable garden in the vicinity of DuPont / Chemours will have an intake just below or at the level of the TDI is located. The uncertainty analysis shows that it is very unlikely that these consumers will have exposure to GenX and PFOA through the consumption of vegetable crops that would lead to negative health effects.

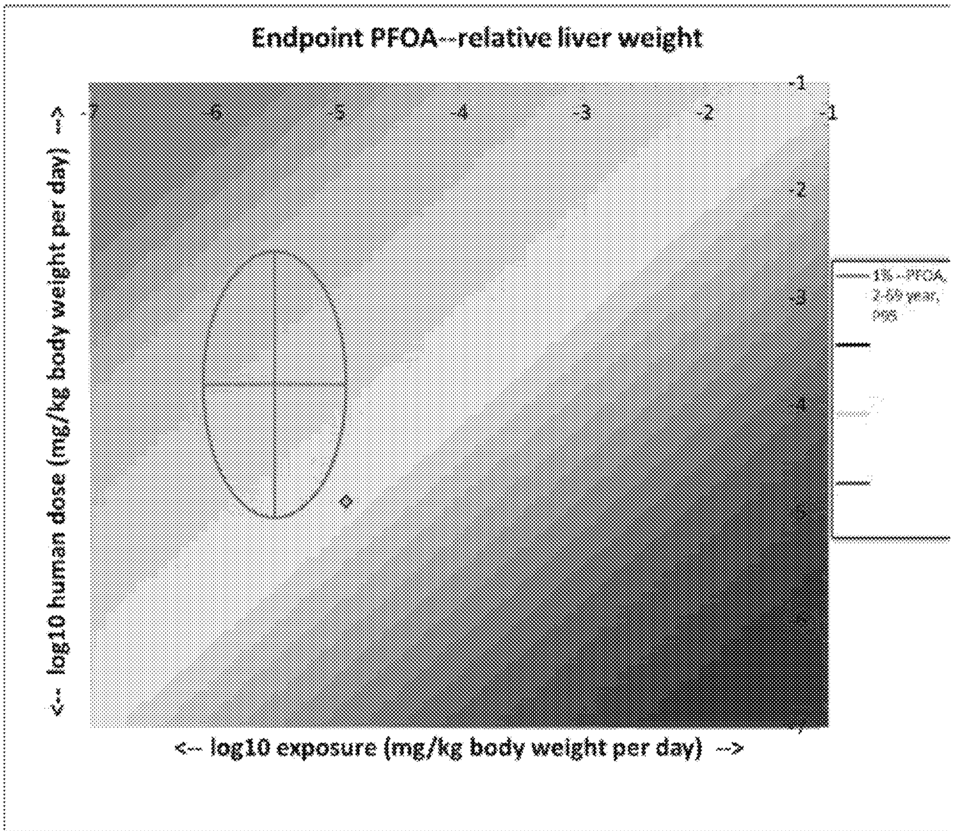

Figure 4. Graphical representation of the uncertainty in the toxicity of PFOA ('human dose', the dose that can cause liver damage in 1% of the consumers, represented by the vertical blue line) and the intake ('exposure', the level of the P95 exposure of consumers, represented by the horizontal blue line) of PFOA through the consumption of vegetable crops

4 Discussion and conclusions

This report describes the risk assessment of GenX and PFOA via the consumption of vegetable crops sampled at various locations around the chemistry company DuPont / Chemours. This chapter discusses the results and draws the conclusions.

4.1 Comparison with previous research related to food

As far as we know, no previous food intake calculations have been performed for GenX. For PFOA, a scientific opinion from EFSA in 2008 states that the median and high intake via drinking water and food was respectively 2 and 6 ng / kg bw / day (EFSA, 2008). The high intake results in a filling of 48% of the current TDI. The most important contribution to this intake was provided by the consumption of fish and fish products.

EFSA did indicate that the intake calculation was based on a deficient set of concentration data in food (EFSA, 2008). Therefore, a survey was conducted for PFOA (and other perfluorinated compounds) in 2009-2010 on the intake by the Dutch consumer based on the consumption of drinking water and food purchased in various supermarkets (Noorlander et al., 2011). To calculate the PFOA intake via drinking water, a concentration of 9 ng PFOA / liter was used, published by EFSA in 2008 (EFSA, 2008). The median (P50) intake of PFOA for drinking water and food in the Netherlands was 0.2 ng / kg bw / day. The high intake (P99) of PFOA was 0.5 ng / kg bw / day. The most important contribution to the intake of PFOA was provided by fruit and vegetables. Based on the current TDI for PFOA of 12.5 ng / kg bw / day, the high intake would mean a filling of the TDI of 4%. The then (median and high) intake is comparable with the (median and high) intake of PFOA in the minimal scenario.

4.2 Found concentrations of GenX and PFOA

GenX and PFOA were not detected in 60% of the samples taken at the sites around the DuPont / Chemours plant (Figure 1). On

GenX could be quantified (concentrations ranged from 1.1 to 5.9 ng / g) and on 3 samples of PFOA (concentrations ranged from 1.3 to 2.8 ng / g). For GenX this concerned samples measured at the locations G1LOC1, G3LOC2 and G3LOC4. For PFOA, quantifiable concentrations were only measured on site G3LOC4.

G3LOC2 and G3LOC4 were located at a distance of less than 1 km from the DuPont / Chemours company (Table 4). G1LOC was located just outside the radius of 1 km. The highest GenX and PFOA concentrations were measured on G3LOC4 and with these concentrations the intake calculations were performed. This means that the intake of GenX or PFOA based on the samples taken at the other locations will always be lower than currently calculated for G3LOC4.

For GenX there was evidence that washing a vegetable led to lower concentrations of GenX. This evidence was not available for PFOA, because PFOA was only found in quantifiable concentrations in three samples of beet. The PFOA concentrations were lower in crops. Subject to the small number of samples, it can be concluded that it is plausible that vegetable washing will reduce the concentrations of GenX and PFOA in vegetables.

In the exploratory study of the VU to the level of GenX and PFOA in leaves and grass in the vicinity of the company DuPont / Chemours, the GenX concentrations ranged from 1.0-28.2 ng / g and for PFOA from 0.4- 19.8 ng / g from 200 to 3000 m. These concentrations are in the same order of magnitude as the concentrations of GenX and PFOA measured in this study. In particular, the concentrations in grass (0.4-5.4 ng / g) correspond well with the concentrations in the vegetable crops. In leaves the concentrations of GenX and PFOA are somewhat higher than in the vegetable crops.

4.3 Uncertainties in the risk assessment

The risk assessment described in this report is based on an indicative study of the intake of GenX and PFOA by consumption of vegetable crops in the vicinity of the DuPont / Chemours plant. In about 60% of the 74 samples, the concentration of GenX or PFOA was below the detection limit. One location showed significantly higher concentrations than the other nine locations around the plant. These concentrations (from 22 samples) were used as a 'worst case' situation to calculate the intake of GenX and PFOA. The intake calculation is based on a minimum and a maximum scenario. In the minimal scenario, the lowest measured concentration per (category) crop was used at this location and in the maximum scenario the highest measured content per (category) crop was used at this location. For GenX, this resulted in a factor of 4 difference between the minimum and maximum scenario for the high intake (P95). For PFOA this factor was equal to 15. The large difference between the minimum and maximum scenario for PFOA was caused by the fact that PFOA could only be quantified in one crop (beets). As a result, the difference between the scenarios was strongly determined by the difference in LOD and LOQ (Table 2). The measured concentrations of GenX and PFOA particularly contributed to the uncertainty in the calculated intakes due to the limited number of samples in combination with a high percentage of samples with a level below LOQ or LOD. The uncertainty in the intake was further influenced by uncertainties in the consumption data used, the effect of washing and the extrapolation of concentrations in non-measured to measured crops.

For the risk assessment, the calculated intake was first set against the allowable daily intake (TDI) for GenX and PFOA, as derived in the recent past by the RIVM. Subsequently, the APROBA-Plus method was used to refine the risk assessment. The analysis with the APROBA-Plus method showed that the largest contribution to the uncertainty in the toxicity of both GenX and PFOA is provided by differences between test animals and humans in kinetics and by the dosage where no negative health effects occur in the test animal, the so-called No Observed Adverse Effect Level (NOAEL). Moreover, the uncertainty analysis also shows that the sum of uncertainties in toxicity is greater than the uncertainties in the intake.

It is therefore obvious to spend any future research on reducing the contribution of the important sources to the uncertainty regarding. the toxicity, ie the interspecies differ in kinetics and the NOAEL. In addition to more clarity about the accumulation of GenX in humans, replacing the NOAEL with a so-called Bench Mark Dose (BMD) can offer a solution. The most cost-effective solution is to derive the BMDs for GenX and PFOA on the basis of the current laboratory animal experiments.

4.4 Exposure from other sources

Other known sources of exposure to GenX and PFOA are drinking water and air. In the calculations reported here for the intake of GenX and PFOA via food, these sources were not included.

In 2016, RIVM estimated the exposure to PFOA via air, drinking water and food in the vicinity of Dupont / Chemours on the basis of emission data, measurement data and calculations (Zeilmaker et al., 2016). The exposure to PFOA via drinking water and food was based on a PFOA concentration in drinking water of 2.5 ng PFOA / liter found in the Dordrecht region in 2015 and the exposure from food in the Netherlands as published by Noorlander in 2011 (Noorlander, 2011). . This resulted in an estimated exposure via drinking water and food between 0.18 (median) and 0.44 (P99) ng / kg bw / day. These intakes are similar to those of Noorlander et al. (2009) via food and drinking water (see section 4.1).

4.4.1 Exposure via air

Zeilmaker et al (2016) did not measure the exposure to PFOA via the air but calculated on the basis of various scenarios for the height and duration of the exposure. The course of the calculated blood serum concentrations showed that, depending on the scenario, there was a high exposure to PFOA in the area within a radius of approximately 750 m from the factory ('inner contour') (with maximum serum concentrations above safe limit of

89 ng PFOA / ml in two of the three scenarios). These calculations assume that no emission of PFOA has taken place after 2012. Because of this emission stop, the calculated serum concentrations in the inner contour in 2016 had dropped to 10 ng PFOA / ml (Zeilmaker et al., 2016). The safe limit of 89 ng PFOA / ml serum corresponds to the TDI of 12.5 ng / kg bw / day. On this basis, a serum concentration of 10 ng PFOA / ml corresponds to chronic air exposure of 1.4 ng PFOA / kg bw / day (= 11.2% of the TDI). This

prehistory also shows that it is very likely that at the end of the last century PFOA was present in higher concentrations in vegetable crops than measured in this study.

In a report from 2016, exposure to GenX was calculated using air (Beekman et al., 2016). Based on the reported emissions in 2014, the estimated exposure for GenX is 15 ng / m3 for the closest inhabited areas (behind the dike on the other side of the river). At a standard tidal volume of 20 m3 air per day for an adult with a body weight of 70 kg (ECHA, 2012) an exposure of 4.3 ng GenX / kg bw / day (= 20.4% of the TDI) was calculated. Time-related exposure to GenX via food can not be concluded because of the snapshot of this study and the lack of other research.

4.4.2 Exposure via drinking water

The 2017 WHO drinking water quality guideline states that for unspecified chemical substances that can occur in drinking water, 20% of the health-based limit value (such as a TDI) is standardized (read: 'reserved') for the exposure via this source (WHO, 2017). Based on this allocation and the TDIs for GenX and PFOA, the RIVM calculated provisional guidance values in drinking water of 87.5 ng / liter for PFOA and 150 ng / liter for GenX (Smit & Versteegh, 2017). Last year GenX was shown in an exploratory measurement program in the Netherlands in the drinking water of three companies (Versteegh & de Voogt, 2017). These companies analyze the surface and bank groundwater downstream from Dordrecht and the Lek (via tidal activities) and from the Meuse (Keizersveer and Brakel). The GenX concentrations were in the range of 10-30 ng / liter. These findings correspond to the highest GenX concentration of 11 ng / liter found in drinking water in the municipalities surrounding the DuPont / Chemours plant (including Dordrecht, Papendrecht and Sliedrecht) by Gebbink et al. (2017).

Based on a daily consumption of 2 liters of drinking water and a body weight of 70 kg, these concentrations lead to a filling of the TDI of 1.4 to 4% for GenX. A concentration in drinking water of 2.5 ng of PFOA / liter, as found in 2015 in the Dordrecht region, leads under the same assumptions for PFOA to a filling of the TDI of less than 1%. In summary, the current limited data set for concentrations of GenX and PFOA in drinking water suggest that the contribution to exposure via this route is very small.

4.5 Contribution exposure via drinking water, air and vegetable crops to total exposure to GenX and PFOA

Table 8 gives an overview of the contributions from the sources of drinking water, air and vegetable crops, at a distance of less than 1 km from the DuPont / Chemours company and for the maximum scenario, to the exposure of GenX and PFOA expressed as a percentage of the TDI. .

The contribution of drinking water to the filling of the TDI is much lower for both GenX and PFOA than for the other sources. For the average intake of GenX and PFOA through the consumption of vegetable crops in the maximum scenario, the contribution of vegetable crops for GenX is higher than the contribution of air (36% versus 20%) and for PFOA more than threefold (34% versus 11%).

Table 8. The contribution of the sources of drinking water, air and vegetable crops, at a distance of less than 1 km from the company DuPont / Chemours and for the maximum scenario, to the exposure of GenX and PFOA expressed as a percentage of the permissible daily intake (TDI)

Dust	Contribution from sources (in % of the TDI[1])			
	Drinkwater average[2]	Air gemiddeld[2]	Kitchen Garden	Total (rounded)
GenX	1,4 - 4	20	Average[3]: 36	57 - 60
			P95[4]: 100	121 - 124
PFOA	0,6	11[5]	Average[3]: 34	46
			P95[4]: 96	108

1 GenX: TDI = 21.0 ng / kg bw / day; PFOA: TDI = 12.5 ng / kg bw / day

2 This exposure is estimated as mean exposure (the distribution of exposure is unknown) (section 4.4.1 and 4.4.2)

3 Average: average exposure in the maximum scenario (Table 7).

4 P95: 95th percentile of the exposure in the maximum scenario (Table 7).

5 The 11% contribution is based on the estimated blood serum value of 10 ng / ml (section 4.4.1).

At a high intake (P95) of GenX and PFOA via vegetable crops in the maximum scenario contributes this source by far the most to exposure (96-100%). In summary, the total contribution of drinking water, air and vegetable crops to the filling of the TDI varied from approximately 57% to 124% in the maximum scenario for GenX and from 46 to 108% for PFOA.

4.6 Summary and final conclusions

Two questions have been formulated for the kitchen garden research into GenX and PFOA:

1. What are the concentrations of GenX and PFOA in selected crops from vegetable gardens in the vicinity of DuPont / Chemours?

2. Is the allowable daily intake (TDI) of GenX and PFOA exceeded by consumption of vegetable crops in a usual consumption pattern?

On question 1, this report provides a response by measuring 74 samples of vegetable crops from 10 locations in the vicinity of DuPont / Chemours. For the answer to question 2, the calculated intake has been compared with the current TDIs of GenX and PFOA. In the maximum scenario, the high intake (P95) was just below (for PFOA) or at the level of the TDI (for GenX).

The aim of this research is to find out whether people with a vegetable garden in the vicinity of DuPont / Chemours can safely consume the vegetables and fruit from this (vegetable) garden with regard to the presence of GenX and PFOA in the crops. The limit values that apply to exposure via food have not been exceeded, and therefore do not in themselves give cause for concern. However, in view of the low contribution of drinking water and the inevitable contribution of air to the exposure, it is advisable to consume vegetable crops that are grown within a radius of 1 kilometer from the farm in moderation (not too often or too much). Outside this area, concentrations are so low that the crops can be safely eaten, also in combination with other exposure sources.

4.7 Recommendations for follow-up research

The results of this study are indicative because of the limited sampling at one point (end of August 2017). It is possible that there is a time-dependent influence (eg seasonal influence) on the concentrations in vegetable crops. Given the very small percentage of samples in which GenX or PFOA could be quantified, it is not expected that this percentage will change significantly in a repeated study. A study with substantially more samples may reduce the uncertainty related to the current concentrations (and therefore to the intake). As already indicated, more profit seems to be gained in reducing the uncertainties related to toxicity.

Research into soil and water samples taken at the current locations (the planned second research phase) will teach us more about the behavior of these substances in the soil and absorption via the crops. However, this knowledge will have little influence on the outcome of these or future risk assessments.

The current measurements, taking into account the limited experimental design, show that PFOA is only found in one crop at one location (less than 1 km away from the plant) at quantifiable levels, which fits into the picture that the use of PFOA by the DuPont / Chemours company stopped in 2012. GenX has been quantified in five plants at two locations (both also less than 1 km away from the plant) and the cause of this is most probably in the GenX emissions from the DuPont / Chemours company. If measurements are desired, it is obvious to measure the exposure to GenX via the air.

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